HOW IMPORTANT ARE MARITIME EMISSIONS FOR THE AIR QUALITY: AT EUROPEAN AND NATIONAL SCALE

A. Monteiro[a*], M. Russo[b], C. Gama[c], C. Borrego[d]

[a]CESAM & Department of Environment and Planning, University of Aveiro, Aveiro, Portugal;

*Corresponding author: e-mail: alexandra.monteiro@ua.pt; Tel: +351 234 370220, Fax: +351 234 370309

[b]CESAM & Department of Environment and Planning, University of Aveiro, Aveiro, Portugal; michaelarusso@ua.pt

[c]CESAM & Department of Environment and Planning, University of Aveiro, Aveiro, Portugal; carlagama@ua.pt

[d]CESAM & Department of Environment and Planning, University of Aveiro, Aveiro, Portugal; cborrego@ua.pt

Abstract

Due to its dependence on fossil fuel combustion, emissions from the marine transport sector can significantly contribute to air pollution. This work aims to evaluate the impact of maritime transport emissions on air quality in Portugal using a numerical air quality modelling approach, with high-resolution emission data. Emissions from the European TNO inventory were compiled and pre-processed at hourly and high spatial (~3x3 km^2) resolutions. Scenarios with and without these maritime emissions were then simulated with the WRF-CHIMERE modelling system, extensively tested and validated for Portugal domain, in order to evaluate their impact on air quality. A simulation was performed for one year (2016) and the resulting differences were analysed in terms of spatial distribution, time series and deltas. The main deltas for NO_2 and PM10 are located over international shipping routes and major ports, while O_3 concentrations are impacted in a larger area. The modelling results also indicate that shipping emissions are responsible for deltas in the concentration of NO_2 higher than 20% over specific urban areas located in the west coast of Portugal, and less than 5% for PM10. For O_3 the relative contribution is low (around 2%) but this contribution is also observed at locations more than 50 km from the coast.

Keywords: atmospheric pollutants; emissions; shipping; air quality modelling; Portugal.

1. INTRODUCTION

Maritime transport is an important sector in Europe that enables trade and contact between all European countries, with almost 90% of the external freight trade being seaborne (Jonson et al., 2015). This form of transportation has been increasing due to the globalization of manufacturing processes and the increase of global-scale trade (Corbett and Fishbeck, 1997; Marmer et al., 2009; Viana et al, 2014). Nevertheless, emissions from the marine transport sector can have a significant impact on atmospheric concentrations of several pollutants (Tsyro and Berge, 1997; Lawrence and Crutzen, 1999; Endresen et al., 2003; Marmer and Langmann, 2005; Wang et al., 2008; Mathias et al, 2010; Gonzalez et al., 2011; Liu et al., 2018), mainly emitting carbon dioxide (CO_2), nitrogen oxides (NOx), sulphur dioxide (SO_2), carbon monoxide (CO), hydrocarbons and primary particulates, as well as secondary particulate precursors (EEA, 2013).

Oceangoing ships have been credited with approximately 15% and 58% of global anthropogenic NOx and SOx emissions, respectively (Eyring et al., 2005; Corbett et al., 2007). Because nearly 70% of ship emissions are estimated to occur within 400 km of land (Endresen et al., 2003), ships can significantly contribute to air quality degradation in coastal areas.

During the last two decades, large efforts have been made in Europe to reduce other types of emission sources (industrial, power generation, etc.), which has increased the weight of shipping emissions relative to total anthropogenic emissions (EEA, 2013). It is therefore important to understand the atmospheric impacts of shipping emissions, especially on regional and coastal air quality.

Corbett et al. (2007) have recently shown that shipping emissions lead to an increase, at a global scale, in air concentrations of fine particles with a diameter less than 2.5 μm (PM2.5), which is linked with premature deaths due to cardiopulmonary diseases and lung cancer (Winebrake et al., 2009). Vutukuru and Dabdub (2008) evaluated the impacts of shipping emissions on tropospheric ozone (O_3) and PM concentrations over California for the first time. Other studies also showed an increase in surface ozone when NOx emissions from ships are included in a global chemistry transport model (Lawrence and Crutzen, 1999). A maximum perturbation of 12 ppb for tropospheric ozone concentrations was found by Endresen et al. (2003) at a global scale. Very

recently, Mertens et al. (2018) estimate that the contribution of shipping emissions to O_3 during summer is up to 20-30%. Besides O_3, Capaldo et al. (1999) calculated an increase of SO_2 concentrations as high as 60% when sulphur emissions from ships are included in a global model. Shipping emissions also lead to an increase in aerosol production through enhancement of OH radical concentration. A 30% increase in sulphate aerosol is predicted due to sulphur emission from ships (Capaldo et al., 1999). Other studies, focused on Asia, pointed out that parts of Southeast Asia receive significant amounts of sulphur deposition, also due to shipping emissions (Streets et al., 2000). Derwent et al. (2005) applied a Lagrangian chemistry-transport model and showed that the contribution of ships to sulphur deposition can reach 55% in some locations in Europe.

Most of the previous studies have dealt with the impacts of shipping emissions at a global and continental scale, using global models with coarse horizontal resolutions (Dalsøren et al., 2007; Collins et al., 2009; Aulinger et al., 2016; Zhang et al., 2017). Relatively few studies considered the impacts of shipping emissions in detail (Eyring et al., 2010; Huszar et al., 2010; Jonson et al., 2015; Aksoyoglu et al., 2016; Johansson et al., 2017). This makes it rather difficult to estimate the effects of shipping emissions on coastal areas and resident population. There is still a lack of knowledge at finer scales, in both recent years (when stricter policies exist for shipping emissions) and including all types of pollutants (primary and secondary, such as ozone, which is particularly critical in southern Europe).

The ongoing AIRSHIP project (http://airship.web.ua.pt/) intends to assess the impact of shipping emissions on the air quality over Portugal and to design effective regulation to minimise the environmental impacts of these emissions. The present work is part of this research project and bridges that gap by conducting a modelling study for Portugal, where there are several large and important ports near urban airsheds. In this study, we investigated the impacts of current ship emissions on the air quality over Europe, and in particular, Portugal. This is achieved using a numerical air quality modelling system, extensively validated, and focusing on the most critical pollutants that are presently exceeding the legislated values over Portugal and Europe (EEA,

2017; Monteiro et al., 2007a), namely: NO_2, PM10 and O_3; in order to obtain an integrated and quantitative picture of these impacts.

The paper is organised as follows. In Section 2, the modelling approach is described in detail. Section 3 focuses on the analysis of the results and the assessment of the air quality impact, per pollutant. Finally, in Section 4, the main conclusions are summarized.

2. THE MODELLING SYSTEM AND ITS SETUP

Numerical modelling has become a fundamental tool to support decision makers on air quality management due to its capacity to estimate atmospheric pollutants concentrations over an entire region, taking into account complex and non-linear physical and chemical mechanisms that characterize the atmosphere, as well as evaluate the effectiveness of emission scenarios. Using sensitivity analysis, these numerical models can be applied to estimate the impact of pollutant concentrations that result from a change in one or more emission sources (Clappier et al., 2017). A mesoscale numerical modelling system was selected and applied in the present study. The effects of shipping emissions on air quality in Portugal was investigated using scenario analysis. Following is a detailed description of the modelling system and its application.

2.1. The WRF-CHIMERE modelling system

The air quality modelling system includes the Weather Research & Forecasting model (WRF version 3.5.0, Advanced Research WRF dynamic solver) (Skamarock and Klemp, 2008) and the CHIMERE chemical transport model (Menut et al., 2013; Mailler et al., 2017) (see Figure 1). The WRF model, developed by the National Center for Atmospheric Research (NCAR), is a next generation mesoscale numerical weather prediction system designed to serve both operational forecasting and atmospheric research needs. CHIMERE is a comprehensive Eulerian air quality modelling system in a non-hydrostatic configuration. Its nesting capabilities enable it to telescope from 1 000 km to 1 km of horizontal resolution, which allows it to combine high grid resolutions, the representation of large-scale transport processes and long-term simulations for emission control scenarios. This modelling system has been extensively used for Europe and, particularly,

Portugal domains (Monteiro et al., 2005; 2007b; Borrego et al., 2011). In addition, it is currently being used as the Portuguese national operational modelling system for daily air quality forecast (http://previsao-qar.web.ua.pt/).

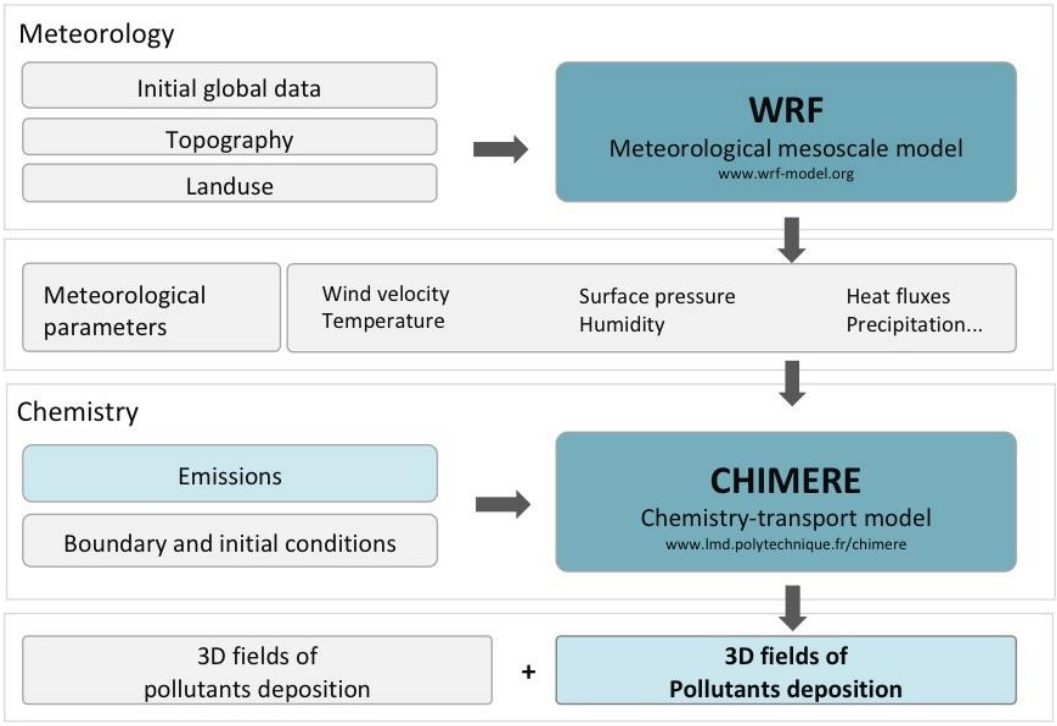


Figure 1. The modelling system WRF-CHIMERE applied in this study.

2.2. Modelling setup and application

The WRF-CHIMERE simulations used three different spatial domains in order to reach a high-resolution scale over Portugal, using nesting capabilities. At first, a grid with a large extent, continental scale, covering southern Europe with a low horizontal resolution of 27x27 km^2 (CONT27, the coarse domain); then a second domain covering the Iberian Peninsula with 9x9 km^2 of horizontal resolution (IP09) and, finally, a high-resolution domain that covers mainland Portugal, with 3x3 km^2 (PT03). These different simulation domains are geographically represented in Figure 2.

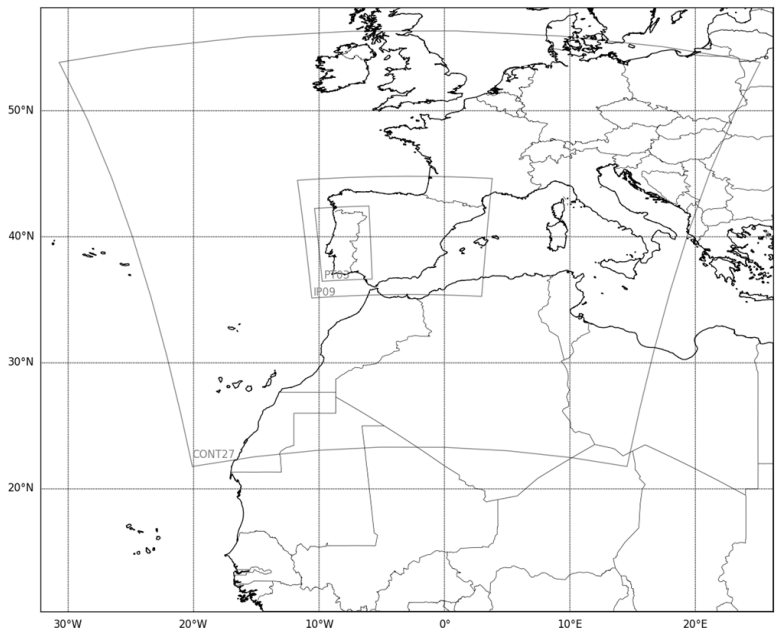


Figure 2. Simulation domains defined for the WRF-CHIMERE modelling application (Horizontal resolutions: CONT27: 27 km^2, IP09: 9 km^2, IP03: 3 km^2).

Regarding the specific WRF and CHIMERE settings for the simulations performed (such as, vertical resolution, parametrizations and boundary conditions), these can be found summarized in Table 1.

The global meteorological fields from the National Center for Environmental Prediction (NCEP/NOAA, 2000), which provide final operational global data on 1º by 1º grids with a temporal resolution of six hours, were used to supply initial and boundary conditions for the coarse domain (CONT27). For the rest, the initial and boundary conditions come from the respective parent domain and from the previous simulated day.

This modelling system was applied for the full 2016 year (the most recent year with available emissions and air quality data), coupled with the most updated and complete emissions inventory available (see Table 1).

Table 1. WRF and CHIMERE models specifications

WRF (version 3.5.0)	

Microphysics	WSM6 scheme (Hong and Lim, 2006)
Cumulus parametrizations	Kain-Fritsch cheme (Kain, 2004)
Planetary boundary layer	ACM2 scheme (Pleim, 2007)
Atmospheric radiation	RRTMG scheme (Iacono et at., 2008)
Grid-nesting techniques	One-way interactive
CHIMERE (version 2016a1)	
Emissions inventory	TNO-MACC_III (0.125º x 0.0625º grid)
Chemistry mechanism	Melchior reduced
Chemically-active aerosols	Yes
Number of aerosol size sections	10
Horizontal and vertical advection schemes	Van Leer I
Number of vertical layers	24
Top layer pressure	200 hPa
Radiative processes	Fast-JX model
Boundary conditions	LMDz-INCA (gaseous and particular species) GOCART (mineral dust)

Emissions data from the TNO-MACC_III inventory were used, as it is the highest spatially resolved emissions source available for Europe (0.125º x 0.0625º gridded) with the complete set of emission sectors as recommended by Russo et al. (2018) in their review paper. This emissions inventory provides anthropogenic emissions data by country and by source category (including shipping), combining the emissions data officially reported by the countries to EMEP, information at country level from the IIASA GAINS model and expert estimates (Kuenen et al., 2014). Regarding shipping activity, Russo et al. (2018) found that the spatial representation shows differences in the emissions distribution, in particular along international shipping routes, as well as Mediterranean, North and Baltic Sea regions. The comparison indicates that TNO emissions are higher over hotspots like the Mediterranean shipping routes, and lower in secondary routes.

The original TNO-MACC emission data was pre-processed for the finest grid resolution, using a spatial disaggregation approach based on land use (see more details in Russo et al., 2018) and temporal disaggregation based on monthly and daily time profiles. The spatial and temporal disaggregation is part of the emission pre-processor of the CHIMERE model

(http://www.lmd.polytechnique.fr/chimere/). Based on this data, Figure 3 shows the contribution of the shipping sector to the total emissions over Europe and Portugal domains.

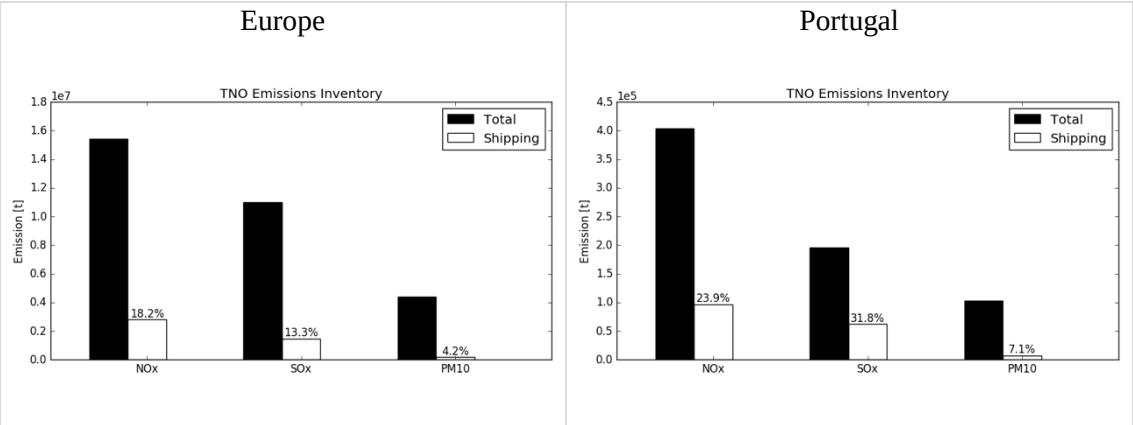


Figure 3. Contribution of the shipping sector to the total emissions for Europe and Portugal domains, for the main primary tropospheric pollutants.

The contribution of shipping activities to total anthropogenic emissions is higher in Portugal than in Europe, with a factor of 1.3 for NOx, 2.4 for SOx and 1.7 for PM10. This was expected due to the geographical location of Portugal and the significant and extensive coastal area of this country. The highest contribution of the shipping sector for Europe is found for NOx (18% for Europe and 24% for Portugal), followed by SOx (13% for Europe, 32% for Portugal) and a smaller value for PM10 (4% for Europe and 7% over Portugal). A higher relative contribution of shipping emissions to SOx emissions in Portugal than in Europe is due to the much lower contribution of other SOx sources in Portugal, especially its very low power plant emissions.

Since shipping emissions are restricted over specific geographical areas (over the international and national shipping routes, close to European coastlines), these contributions (assessed in global terms) in a similar analysis focused on coastal areas.

2.3. Modelling evaluation

This modelling system has already been fully validated for different type of applications and study domains (e.g. Borrego et al., 2011; Monteiro et al., 2013). Nevertheless, a brief model validation

exercise is presented following in order to guarantee the adequate performance of the modelling system in simulating the different pollutants studied. The model outputs for the different pollutant concentrations were compared with observed data measured at different rural and urban background air quality monitoring stations distributed along Portugal.

Figure 4 presents the time series plots of modelled and observed NO_2, PM10 and O_3 concentrations (hourly average concentration for NO_2, daily average concentration for PM10 and daily maximum 8-hour average concentration for O_3). The concentrations presented correspond to the mean values modelled and observed in 6 urban background stations and in 3 rural background stations, i.e., assuming pairing in time but not in space. Common model quality indicators, such as Pearson correlation coefficient (Pearson's r), root mean square error (RMSE) and bias have been calculated (see Table 2).

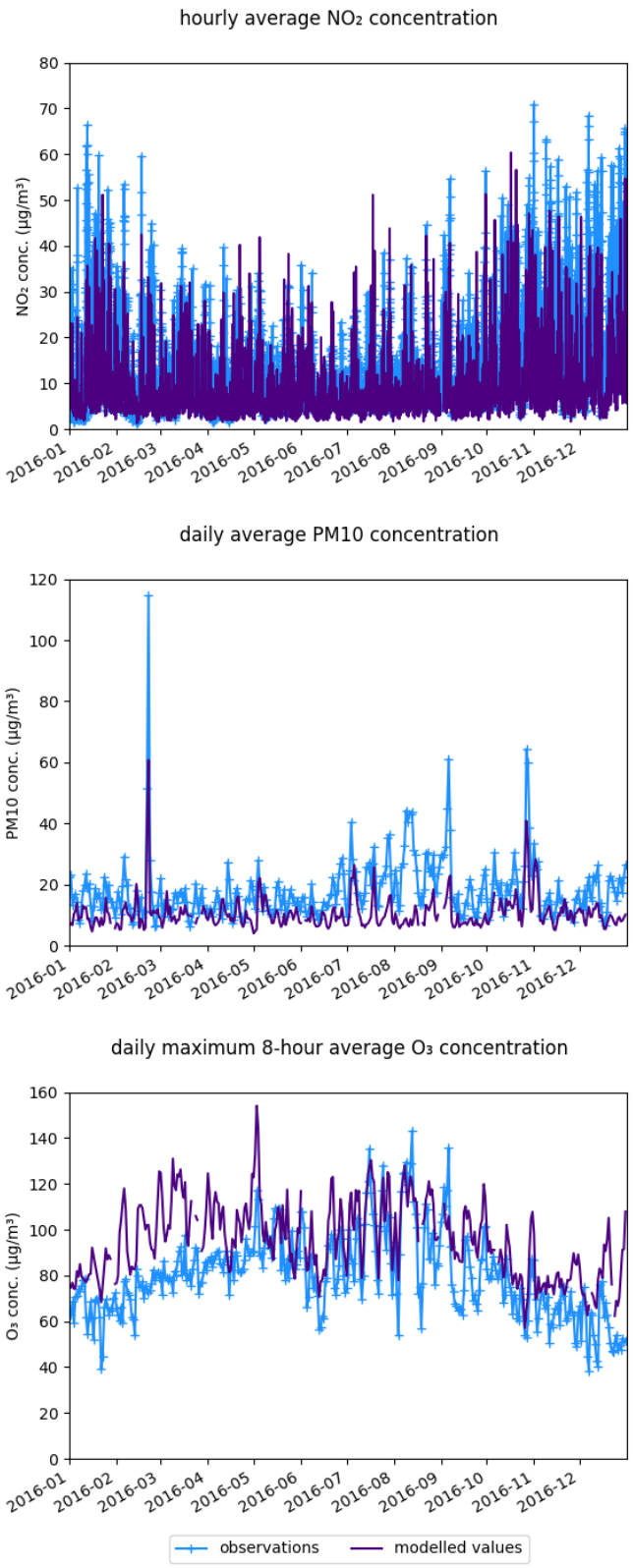


Figure 4. Comparison between modelled (purple) and observed (blue with symbol) NO_2, PM10 and O_3 concentrations in 9 rural and urban background air quality monitoring sites distributed along Portugal.

Table 2. Model quality indicators calculated with average concentrations in 9 sites background sites (6 urban and 3 rural), considering pairing in time but not in space.

Indicators	NO_2	PM10	O_3
Pearson's r	0.73	0.70	0.74
RMSE ($\mu g.m^{-3}$)	7.8	10.4	20.1
bias ($\mu g.m^{-3}$)	-4.1	-7.4	15.7

Good correlations (Pearson's r > 0.7) are observed between modelled and observed series unpaired in space, for the three pollutants. Model systematic under estimates PM10 concentrations (bias = -7.4 $\mu g.m^{-3}$). In Figure 4, several anomalous episodes of high PM10 concentrations are observed, which according to Gama (2018) are due to Saharan dust outbreaks (on February, September and October) and to the influence of numerous wildfires (on the first half of August and the beginning of September). Model simulations take into account mineral dust emissions over North Africa, however their contribution to particulate matter concentrations over Portugal is under estimated. Wildfire emissions are not taken into account by the model. These natural contributions to PM10 levels contribute to a decreased performance of the model. Despite the good correlation calculated for O_3 (Pearson's r = 0.74), the model over estimates observed concentrations between November and April, causing a bias of 15.7 $\mu g.m^{-3}$. During summer, however, particularly during specific episodes, the model is not able to simulate the highest observed concentrations, which may be related with the lack of wildfires emissions.

Overall, the model shows an adequate performance in simulating NO_2, PM10 and O_3 concentrations in the atmosphere, which validates its use in this study.

3. IMPACT OF SHIPPING EMISSIONS ON AIR QUALITY

In order to evaluate and quantify the contribution of the emissions of the shipping sector on air quality, the modelling results for the different scenarios (with and without shipping emissions). The analysis are made in terms of spatial distribution (delta analysis) and regarding time series,

for specific locations, and for the most critical tropospheric pollutants with yearly exceedances over Europe (EEA, 2017) – NO_2, PM10 and O_3.

3.1. Spatial analysis

Figure 5 shows the spatial distribution of differences (mean and maximum deltas) found between the hourly simulations with and without maritime emissions, for the European domain and for the 3 pollutants (NO_2, PM10 and O_3).

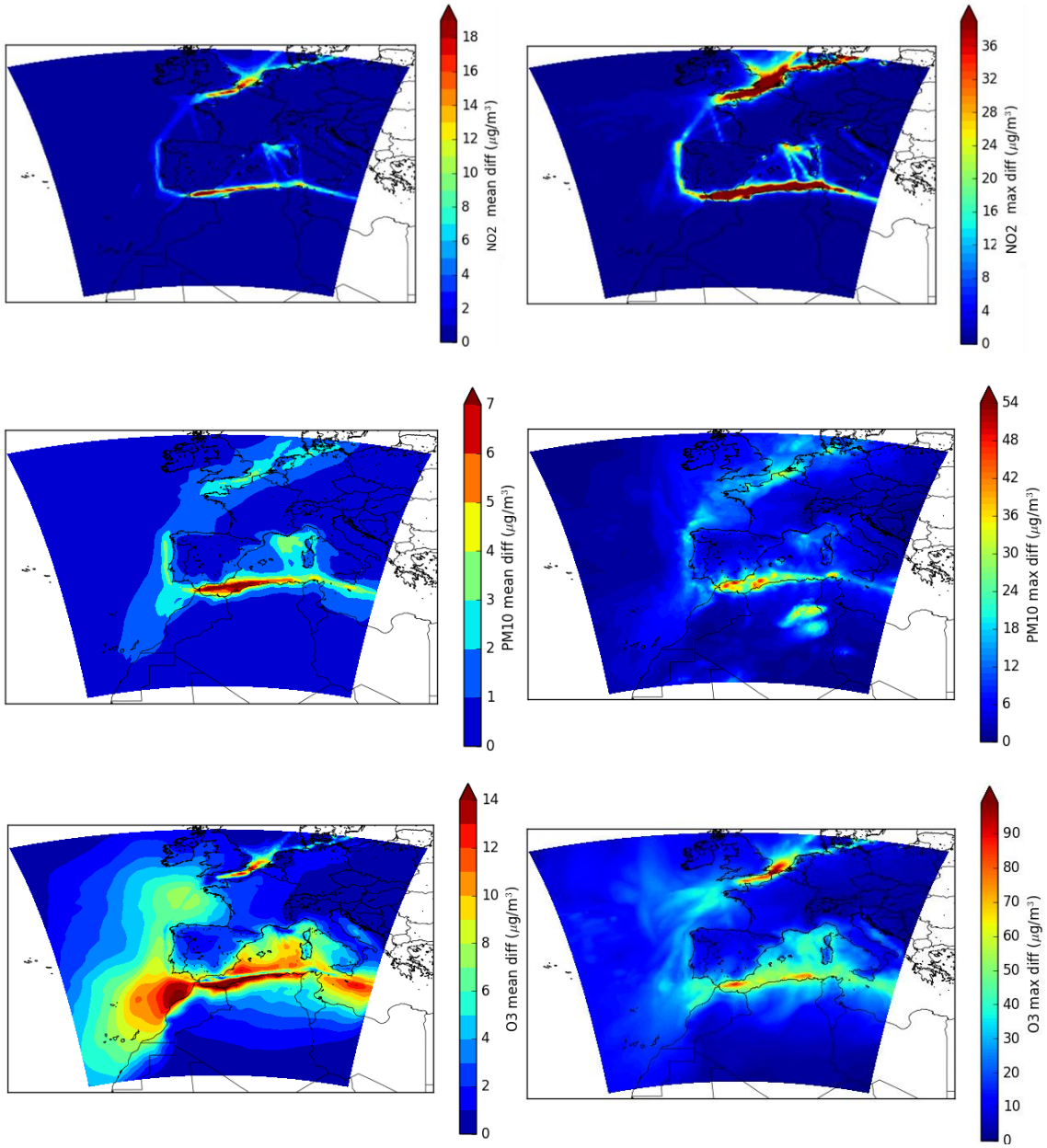
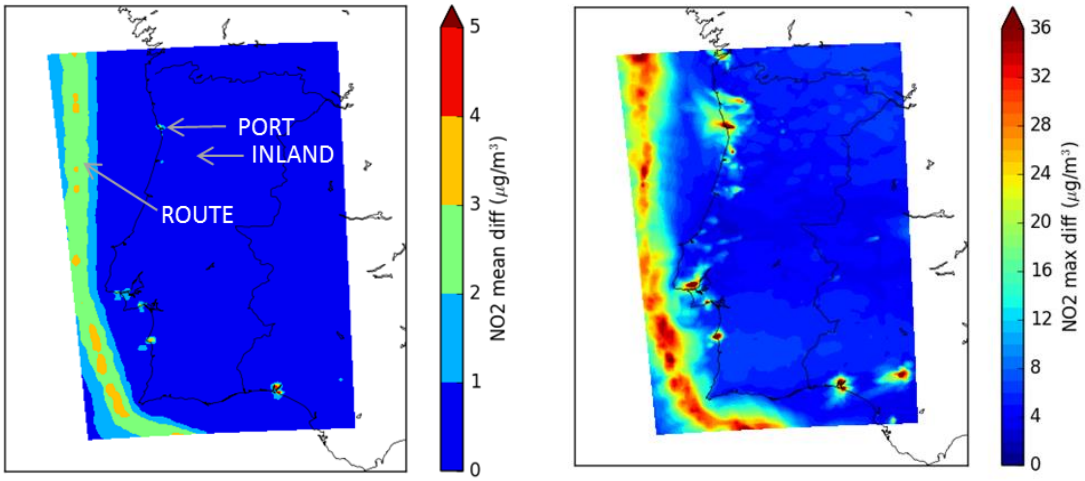


Figure 5. Mean deltas (left) and maximum deltas (right) between the simulations - with and without shipping emissions - for NO_2, PM10 and O_3 concentrations, for the European domain.

Over the Europe, there are two regions where the difference is maximum (with maximum deltas above 35 µg.m^{-3} for NO_2; 50 µg.m^{-3} for PM10 and 80 µg.m^{-3} for O_3), the Mediterranean and North seas. Regarding the mean differences, they reach 18 µg.m^{-3} for NO_2, 7 µg.m^{-3} for PM10 and 14 µg.m^{-3} for O_3, evidenced in the main shipping routes of Straits of La Mancha and Gibraltar. These mean deltas are of the same order of magnitude as the annual average concentrations observed for the primary pollutants, at the same locations over the European domain (EEA, 2017): NO_2 < 20-25 µg.m^{-3} and 15-35 µg.m^{-3} for PM10, showing the significance of shipping emissions in both regions.

Figure 6 shows a similar analysis for Portugal domain.



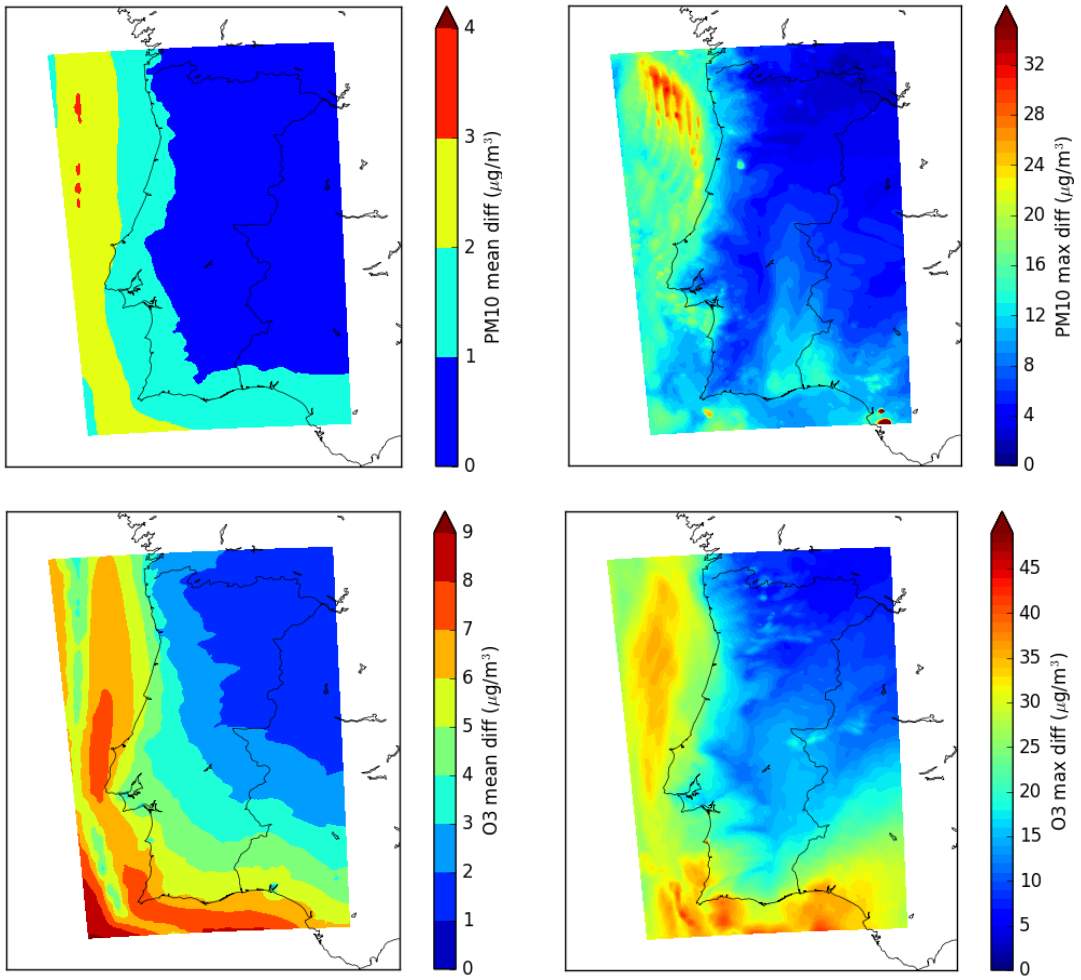


Figure 6. Mean deltas (left) and maximum deltas (right) between the simulations - with and without shipping emissions - for NO_2, PM10 and O_3 concentrations, for Portugal domain.

At this scale, the differences for NO_2 affect a larger area, and are not entirely restricted to international shipping routes and major ports. In the case of PM10, the impact of shipping emissions is noticed over the coast as well as 40-60 km inland. The impacted area on O_3 concentrations is even larger, extending over the entire west and south coast with deltas higher than 8 µg.m^{-3} (on average) and 30 µg.m^{-3} (maximums).

The differentials in terms of NO_2 are in the range 5-20 µg.m^{-3}, with maximums higher than 30 µg.m^{-3}, in the Mediterranean sea. For PM10 the shipping contribution is, on average, only 2-7 µg.m^{-3} but with maximums of 30 µg.m^{-3}. For O_3 the contribution of shipping emissions is approximately 5-15 µg.m^{-3} (with maximums > 60 µg.m^{-3}) in the Mediterranean Sea.

3.2. Time series at different sites

Figures 7, 8 and 9 show hourly, daily and weekday time series simulated with and without considering shipping emissions, for the three studied pollutants (NO_2, PM10 and O_3), at three different locations: along the international route (ROUTE), in a harbor area (PORT) and at a rural inland location (INLAND). These locations are shown in Figure 5. The time series are for a 1-year simulation.

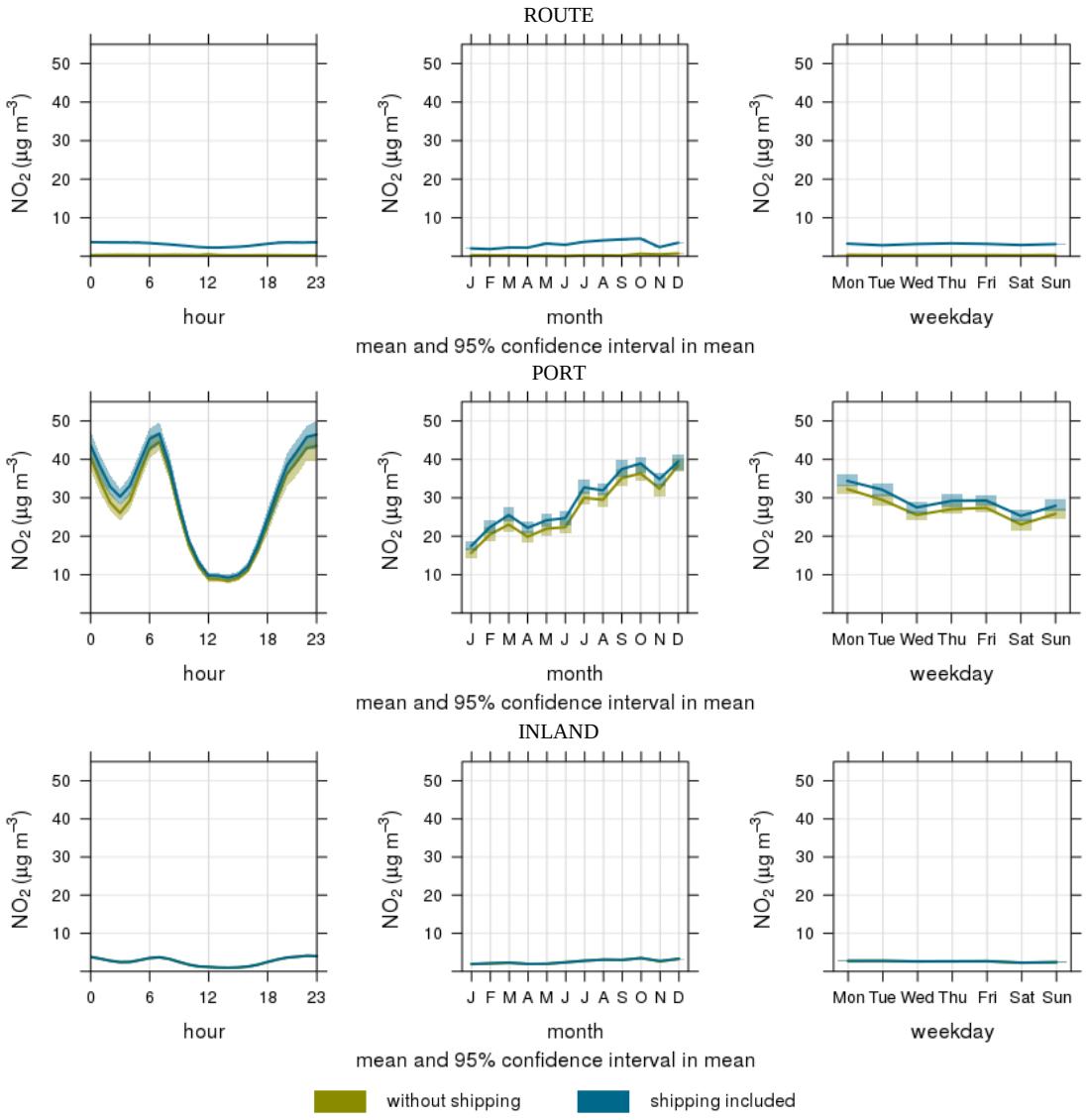


Figure 7. Hourly, daily and weekday time profiles for NO_2 simulated with and without shipping emissions, for three different locations (ROUTE; PORT and INLAND).

ROUTE

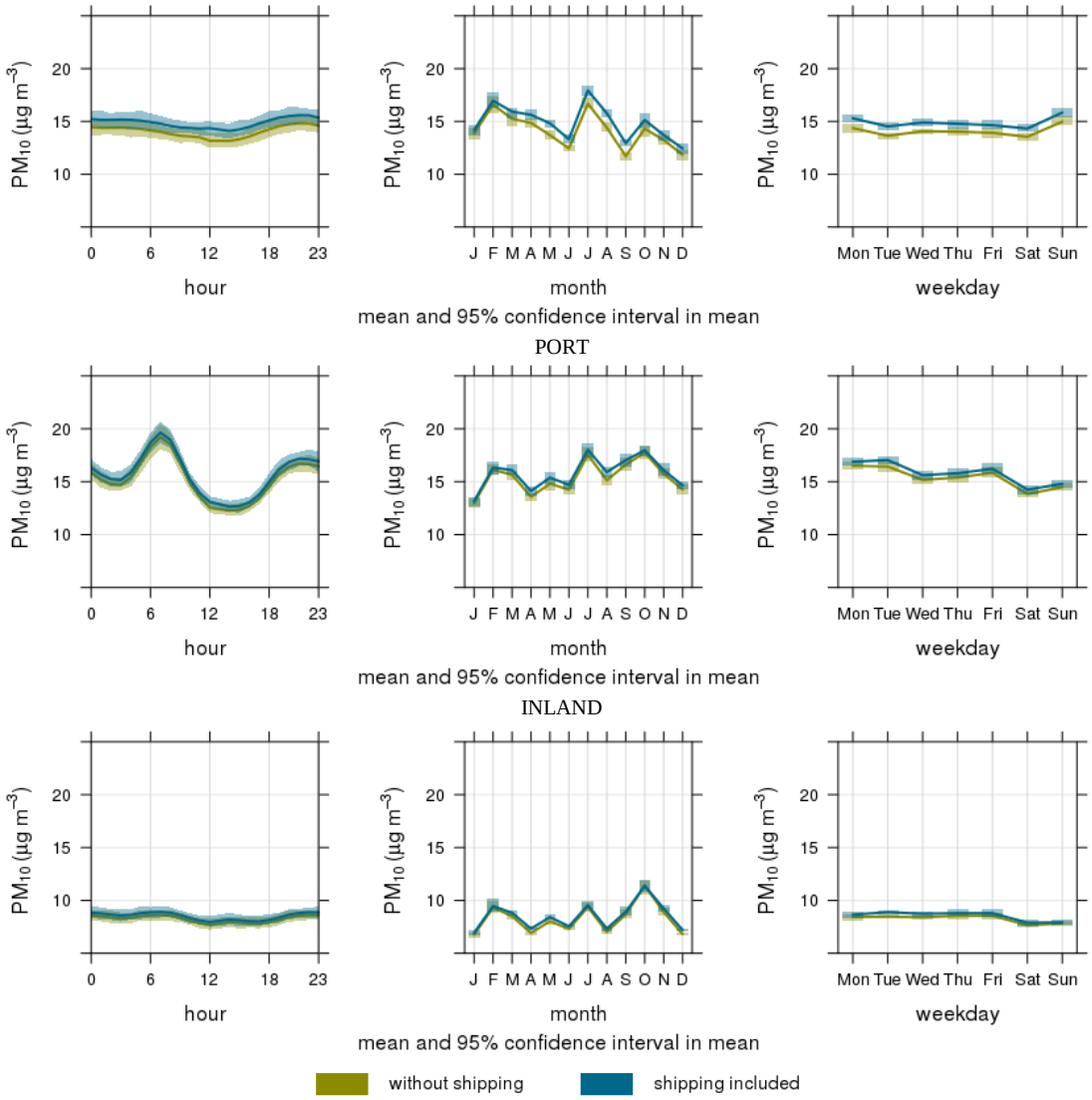
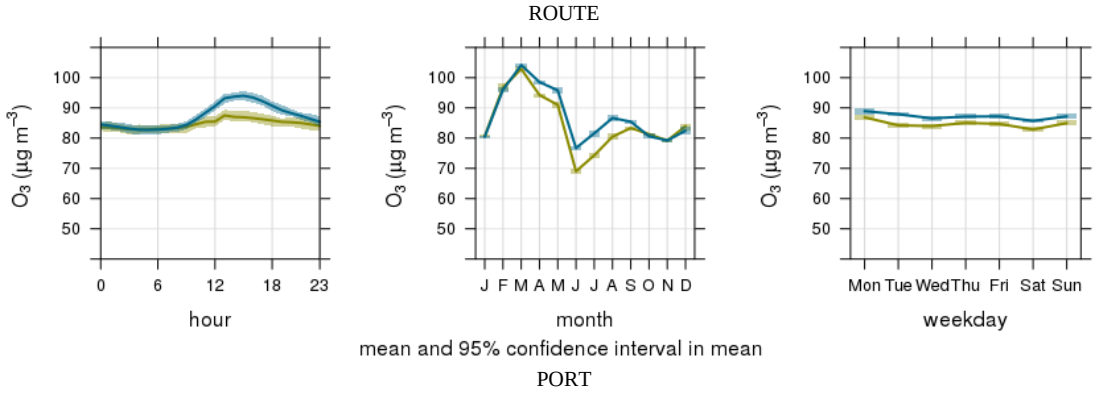


Figure 8. Hourly, daily and weekday time profiles for PM10 simulated with and without shipping emissions, for three different locations (ROUTE; PORT and INLAND).



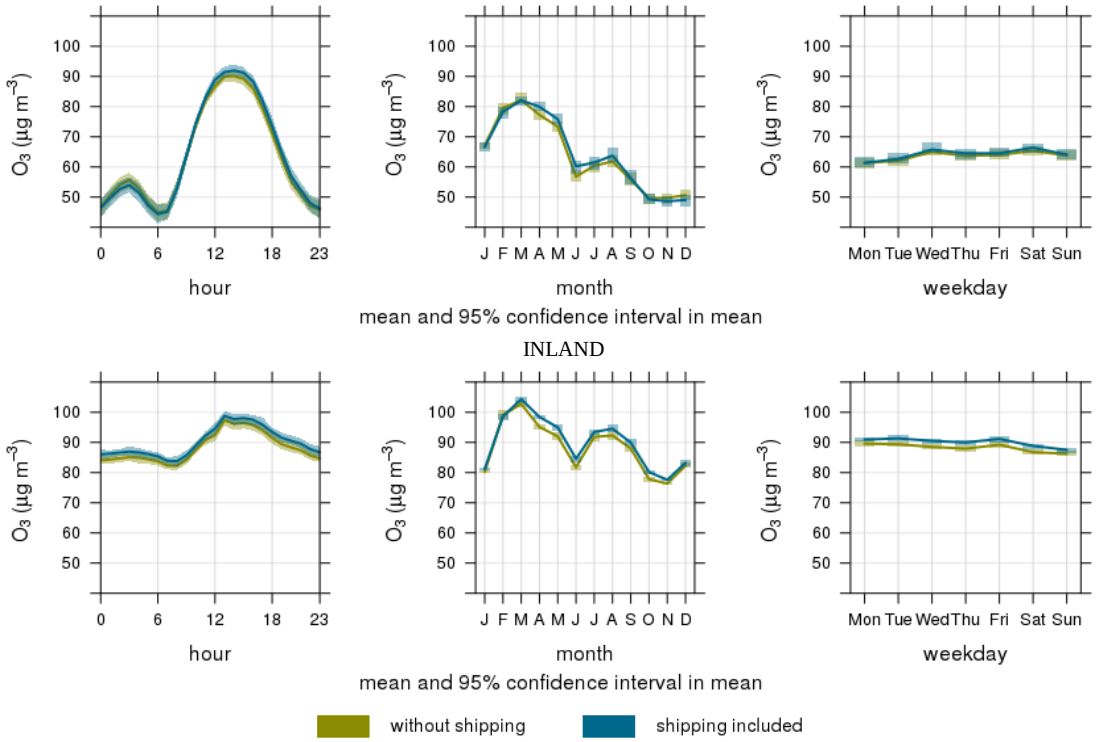


Figure 9. Hourly, daily and weekday time profiles for O_3 simulated with and without shipping emissions, for three different locations (ROUTE; PORT and INLAND).

Regarding NO_2, the contribution of shipping over the ROUTE is absolute, as expected, since it is the only anthropogenic NO_2 source over these areas. Shipping is responsible for NO_2 hourly and daily average values of 4 µg.m^{-3}. Its contribution over the PORT is around 4-5 µg.m^{-3} while INLAND is below 2 µg.m^{-3}. NO_2 absolute values over these two locations – PORT and INLAND – are very different (15-70 µg.m^{-3} for PORT and 2-5 µg.m^{-3} for INLAND), and the the relative contribution of the shipping is 10-20% over PORT and below 5% INLAND. The daily time profile of NO_2 (and PM10) for PORT and INLAND locations confirm the different influence of shipping emissions between them: over the PORT a clear traffic/urban profile is observed (with peaks in the morning and late afternoon) which is not observed in INLAND daily profile.

For PM10, the modelled results indicate that shipping is responsible for concentration values around 1 µg.m^{-3} over the ROUTE. Contributions below 0.5 µg.m^{-3} are expected at the PORT and lower than 0.2 µg.m^{-3} at the INLAND rural location. All these absolute deltas correspond to relative contributions of the shipping activity of around 3%.

Finally, for O_3 an average delta of 1-3 μg.m^{-3} is estimated over the ROUTE, with maximums of 5 μg.m^{-3} in the summer (and during the 12-15 UTC period). A difference below 1 μg.m^{-3} is found in the PORT and close to 2 μg.m^{-3} at the INLAND location (60 km from the coast). The higher delta estimated away from the shipping route (ΔINLAND > ΔPORT) is explained by the secondary origin of O_3, which is formed through its NOx precursors reactions during their transport and dispersion. These absolute deltas correspond to relative contributions of 2%.

4. SUMMARY AND CONCLUSIONS

This study investigates the impact of shipping emissions on the air quality over Europe and, in particular, Portugal. A numerical modelling approach was used to simulate this impact, considering the most critical pollutants in terms of atmospheric concentration over these regions: NO_2, PM10 and O_3. Emissions from the TNO inventory were pre-processed for the defined modelling resolution (3x3 km^2 at the finest domain for Portugal). Modelling results point out that the most critical areas in Europe occur in the North and Mediterranean Seas, limited to the international route. The main differences for NO_2 and PM10 are located over the international shipping routes and major ports. The impacted area on O_3 concentrations is larger, extended over the entire west and south coast with deltas higher than 8 μg.m^{-3} (in average) and 30 μg.m^{-3} (maximums). Over Portugal domain there are deltas of 5-20 μg.m^{-3} for NO_2, with maximums higher than 30 μg.m^{-3}, in the Mediterranean sea. For PM10 the shipping contribution reaches only 2-7 μg.m^{-3} but with maximums of 30 μg.m^{-3}. For O_3 the contribution of shipping emissions is about 5-15 (with maximums > 60 μg.m^{-3} in the Mediterranean Sea).

Pollutants concentrations were analysed at three different sites (at the shipping ROUTE, near a PORT, and at an INLAND area 60 km from the coast). Shipping is responsible for hourly and daily averaged increments of NO_2 around 4 μg.m^{-3}. The relative contribution of the shipping is 10-20% over the PORT location and below 5% at INLAND. For PM10, the modelled results indicate that shipping is responsible for concentration increments around 1 μg.m^{-3} over the ROUTE location. Contributions below 0.5 μg.m^{-3} are expected at the PORT location and below 0.2 μg.m^{-3} at the INLAND rural location, which correspond to relative contributions of

approximately 3%. Finally, for O_3, an averaged delta of 1-3 $\mu g.m^{-3}$ is estimated over the ROUTE location, with maximums of 5 $\mu g.m^{-3}$ in the summer. A difference below 1 $\mu g.m^{-3}$ is found at the PORT location and closer to 2 $\mu g.m^{-3}$ at INLAND (60 km from the coast), which is justified by the secondary nature of this pollutant.

These modelling results will aid the management and political actions in the maritime transport sector and its environmental impacts, specifically concerning the maritime transport and port sectors. In the future, plans will include the study of these impacts considering climate change scenarios.

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